

Identifying and quantifying transported vs. local sources of New York City PM_{2.5} fine particulate matter air pollution

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Abstract

New York City (NYC) is presently in violation of the nation's PM_{2.5} annual mass standard, and will have to take actions to control the sources contributing to these violations. This paper seeks to differentiate the impact of long-range transported aerosols on the air quality of downtown NYC, so that the roles of local sources can more clearly be evaluated. Past source apportionment studies have considered single sites individually in their source apportionment analyses to identify and determine sources affecting that site, often finding secondary sulfates to be an important contributor, but not being able to quantify the portion that is transported vs. local. In this study, a rural site located in Sterling Forest, NY, which is near to the NYC area, but unaffected by local NYC sources, is instead used as a reference to separate the portion of the aerosol that is transported to our Manhattan, NYC site before conducting the source apportionment analysis. Sterling Forest is confirmed as a background site via elemental comparisons with NYC during regional transport episodes of Asian and Sahara sandstorm dusts, as well as by comparisons with a second background site in Chester, NJ. Two different approaches that incorporate Sterling Forest background data into the NYC source apportionment analysis are then applied to quantify local vs. transported aerosols. Six source categories are identified for NYC: regional transported sulfate, trans-continental desert dust, traffic, residual oil, "local" dust and World Trade Center fires pollution. Of these, the transported sulfates and trans-continental desert dust accounted for nearly half of the total PM_{2.5} mass in Manhattan during 2001, with more than half coming from these transported sources during the summer months. More than 90% of the Manhattan elemental carbon was found to be of local origins. Conversely, roughly 90% of the NYC sulfate mass was identified as transported into the city. Our results indicate that transported pollution has a major role in NYC's fine PM pollution. Reliably meeting the ambient PM_{2.5} air quality standards in New York will require that upwind sources, outside of the city, will also need to be controlled.

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1. Introduction

Chemically non-specific particulate matter (PM) mass concentrations cannot alone provide a complete indication of PM toxicity. Instead, it is a combination of size and chemical constituents of

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PM that are proving to be important. In 2000, the US Environmental Protection Agency (EPA) Speciation Trend Network (STN) was initiated to monitor for $\text{PM}_{2.5}$ (particulate matter less than $2.5\ \mu\text{m}$ in diameter) and its elemental constituents at over 200 sites across the US (US EPA, 1999). This provides greater details about the characteristics of PM and using source apportionment tools will allow the identification and estimation of mass contributions from different PM sources (Cooper and Watson, 1980). For a city, such as New York City (NYC), where the impact of “long-range” transport aerosols is well established, speciation data provides an opportunity to apportion NYC $\text{PM}_{2.5}$ into contributions from local and non-local sources.

In the past, multiple studies have compared PM mass and elemental tracers, such as NH_3 , K, NO_3 , SO_4^- , carbon, and H^+ levels, between sites in the eastern US in order to estimate the impacts of secondary and transported aerosols (Vukovich and Sherwell, 2002; Bari et al., 2003; Dutkiewicz et al., 2004; Khan et al., 2006). The availability of multiple years of STN data, has led to several newer source apportionment studies, allowing the identification of the various sources impacting air quality at these sites, and a comparison of the source’s relative impacts. Two recent studies conducted for NYC (Ito et al., 2004; Qin et al., 2006) used every 3-day STN data for multiple sites in the NYC metro area, and conducted independent source apportionment analyses for each of the sites. The types of sources and mass contributions were compared between the sites, and a “secondary sulfate” aerosol source was observed at each of the multiple sites. However, it is unclear exactly how much of the secondary sulfate identified at each site was transported versus local.

In this paper, we investigate an alternate approach to conducting a source apportionment wherein data from an additional “background” site is incorporated into the NYC site source apportionment analysis to help quantitatively estimate the roles of transported vs. local aerosols as a part of the source apportionment. We apply daily elemental data collected during 2001 by New York University (NYU) at two sites located: (1) in downtown Manhattan; and, (2) in Sterling Forest (Tuxedo, NY). Sterling Forest is a rural site surrounded by thousands of acres of largely undeveloped woodland, with an area approximately the size of Manhattan, and it is situated approximately 50 miles west-northwest of NYC. This site was chosen

with the goal of comparison with aerosols collected at the Manhattan site. Sterling Forest experiences the same transported aerosols as in NYC, but none of the local sources. In the absence of unique tracers for the local vs. transported portions of each of the different source contributions, we propose incorporating the complementary data from the background (Sterling Forest) site directly into the NYC source apportionment model in order to provide a quantification of the transported aerosol contribution as an integral part of the NYC site apportionment.

Two approaches that incorporate background data into the model are applied in this research. Since numerous past studies have shown a major impact of transported sulfates into NYC air quality, Sterling Forest sulfur data is included in the NYC source apportionment as an additional variable that should help more clearly define “transported aerosols” in the first apportionment approach. That is, the sulfur measurements taken at the background site would essentially act as a tracer for aerosols transported into the NYC metro area, with the remainder of the NYC sulfur (i.e. the difference in sulfur levels between the two sites) then being more clearly attributed by the model to local sources in the source apportionment analysis. In the second approach, we assume Sterling Forest is a good background site for all transported aerosols (not just sulfates), and therefore it incorporates all the background data into the NYC source apportionment model. In this second approach, two separate source apportionments are conducted on: (1) the Sterling Forest data; and (2) the “remainder” elemental data at NYC after “subtracting off” the Sterling Forest concentrations, in order to then provide separate apportionments for the transported vs. local sources affecting the NYC site.

2. Methods

2.1. Data collection

NYU set up two PM sampling sites in late 2000, and began sampling $\text{PM}_{2.5}$ and its chemical constituents on a daily basis in January 2001. At both sites, 24-h (midnight-to-midnight) $\text{PM}_{2.5}$ filter samples were collected using an R&P ACCU sampler (operating at 13.7 LPM), and every half-hour continuous real-time $\text{PM}_{2.5}$ mass concentration data were simultaneously collected using an R&P TEOM (Thermo Electron Corporation, 2005).

An R&P 5400 carbon analyzer using “thermal- CO_2 ” was also used to measure elemental carbon (EC) and organic carbon (OC) levels every 3 h at each site. The temperatures for the OC and EC were 340 and 750 °C, respectively. Routine flow audits were done for all the R&P instruments.

The samples and blanks were subsequently analyzed for 34 trace element concentrations using energy dispersive X-ray fluorescence (ED-XRF) techniques (Model EX-6600 –AF, Jordan Valley; spectral software XRF2000v3.1 by USEPA and ManTech Environmental Technology, Inc.). Each XRF batch of filters included three to six blanks. Using the EPA XRF program, Spectral Software XRF 2000v3.1 (USEPA and ManTech Environmental Technology, Inc), the elemental concentrations and their associated uncertainties were computed for each daily $\text{PM}_{2.5}$ sample. The program includes the error propagation of calibration uncertainty, long-term system stability, uncertainty in least squares fit, uncertainty in attenuation correction, uncertainty in interference correction, uncertainty in flow rate and the uncertainty in sample deposit area.

2.2. Data (quality assurance)

After filters were weighed and analyzed for the chemical constituents, the daily PM data were aggregated from the various instruments. The data were first screened for days with sampling anomalies, such as power failure or targeted flow rates not being achieved. From the list of 34 elements analyzed by the XRF, 22 were selected for inclusion in the source apportionment analyses based on their concentrations relative to their respective levels of detection. Blanks run through the XRF were also checked for elemental concentrations, as quality assurance. Elemental concentrations of blanks were looked at by individual batch as well as compared between batches over the entire XRF analysis period, to check for XRF detector issues and instrumental drift. Of the 22 elements chosen, only selenium showed drift. There were three batches in the latter half of all the analysis that were found to have higher selenium on the blanks, and therefore the selenium filter concentrations were adjusted by subtracting the blank Se value from the filter values for these batches. The daily EC and OC concentrations were averaged using data from the every-3 h cycles. Days with less than 6 out of 8 complete 3-h cycles were removed from the data set. A standard-

dized 5% of the concentration was used as the carbon uncertainty, as per the recommendation of the manufacturer. For a short period in September 2001, the R&P 5400 had problems attaining the desired temperatures during its analysis cycle at our site located at Hunter College on First Avenue in Manhattan. This missing carbon data has been replaced with an average carbon value of the month before and after this period, with increased uncertainties. This allows us to use all our elemental data for the entire sampling period in the analyses. In this work, data from only 2001 (January 18th–December 27th) are used.

EPA STN elemental data from Chester NJ is limited for 2001 (some 50 observations for the entire year) and therefore is utilized in this work, with the sole purpose of verifying the assumption of Sterling Forest as a suitable background site. EPA gaseous pollutant data has also been used in this work, to aid the confirmation of identified sources. Wind direction data and the on-line HYSPLIT Model from the National Oceanic and Atmospheric Administration (NOAA) were used for supporting MET data analysis.

2.3. Data analysis

2.3.1. Source apportionment tools

Positive matrix factorization (PMF) was used to conduct our source apportionment of NYC elemental data. This technique developed by Paatero (1997), is based on factor analysis and, assumes that x_{ij} i.e. the measured chemical species detected on a given sample is from p independent pollution sources:

$$x_{ij} = \sum_{k=1}^p g_{ik} f_{kj} + e_{ij}$$

(x_{ij} is the j th species concentration measured in the i th sample; g_{ik} is the mass contribution from the k th source on the i th sample; f_{kj} is the j th species mass fraction from the k th source; and, e_{ij} is the error term).

PMF uses a weighted least-squares fit, where weights are based on the uncertainties of the elemental concentration measurements (i.e. u_{ij} is the uncertainty estimate in the j th element measured on the i th sample). Unlike other source apportionment models, this allows for the inclusion of measurement “uncertainties”, thereby providing information regarding the confidence in the concentration

measurements into the model fit, which is one of the major advantages of this method. Further, by including non-negative constraints in the model (i.e., that mass contributions or source profile composition fractions cannot be less than 0), only physically plausible solutions are allowed. Numerous source apportionment studies in recent years have successfully used PMF (e.g., Kim and Hopke, 2004; Song et al., 2001) and more details regarding this technique are available in these published papers.

In this paper, instead of applying PMF to the raw NYC elemental concentration data, we consider two alternate approaches toward estimating the local vs. transported contributions to $PM_{2.5}$ in NYC. These involve incorporating the Sterling Forest background data into the NYC source apportionment analysis. In these methods, we do not assume that the Sterling Forest site is necessarily directly upwind of NYC on a given day; merely that it is similarly impacted by the air pollution being transported into the NY metro area, but not at all by the local NYC pollution. The first uses only the Sterling Forest sulfur variable in the analysis as a tracer of the transported aerosols impacting Sterling Forest and NYC. Therefore, in addition to the usual 20 elements and the two carbon variables (i.e., OC and EC) for the NYC site, two sulfur variables were included as a part of this analysis (a “transported sulfur” variable and a “local sulfur” variable), such that for this Case 1:

$$S_{\text{TRANSPORTED}} = S_{\text{SF}},$$

$$S_{\text{LOCAL}} = S_{\text{NYC}} - S_{\text{SF}},$$

On some days, Sterling Forest sulfur concentrations were found to be slightly higher than in the city, and the NYC S_{LOCAL} was set = 0.0 for those days, in order to avoid negative “local” sulfur concentrations.

The second approach (Case 2) incorporates all Sterling Forest elemental data to separate the local vs. transported elemental concentrations experienced at the Manhattan site. Here, we assume that Sterling Forest is also a good background site for all transport related fine particles, and apply the above approach to all elements (not just sulfur) prior to conducting the NYC PMF source apportionment.

Using these approaches, we seek to use the wealth of information available at Sterling Forest to further enhance our Manhattan apportionment analysis by separating the local and transported

portions of all the elemental concentrations before conducting the PMF. Thus, it would be useful to also use this differencing technique for all the elemental variables to create a separate set of NYC elemental variables consisting of just the remainder or “local” contributions of each of the elements and carbon for application to PMF, separate from a PMF of the transported aerosols, based on the Sterling Forest PMF. The Sterling Forest data thus supplements this new data set with information regarding the regional concentrations. In this case, separate source apportionment analyses were independently conducted on both these two datasets to determine “local” and “transported” sources. Factor scores from each these two analyses were then applied simultaneously in a multiple linear regression of the Manhattan $PM_{2.5}$ mass on all of the identified source components, in order to convert the scores to actual mass concentration estimates for each of the transported and local source components identified by the two separate PMF analyses.

The uncertainties of elemental concentrations used in the PMF analyses are reported by the XRF. The Sterling Forest uncertainties were used for the elemental variables classified as “transported”. For the “local” elemental variables calculated by differencing the NYC and Sterling Forest concentrations, uncertainties were propagated as,

$$U_{\text{LOCAL}} = \sqrt{(U_{\text{SF}}^2 + U_{\text{NYC}}^2)}.$$

3. Results

3.1. Evaluating sterling forest as a NYC background site

Before applying the Sterling Forest data as a background pollution site for NYC, we first evaluated this assumption. To test the suitability of Sterling Forest as a background site, the elemental and carbon data collected at the site were compared with both an STN “background” site located in Chester NJ and NYU’s Manhattan NYC site (a map indicating the location of these sites is provided in Fig. 1). It is apparent from the results in Table 1 that, while $PM_{2.5}$, sulfur and OC are highly correlated across the three sites, EC is far less correlated. This indicates that sulfate and OC are useful tracers of regional transported and secondary aerosols, while EC is a tracer of more local

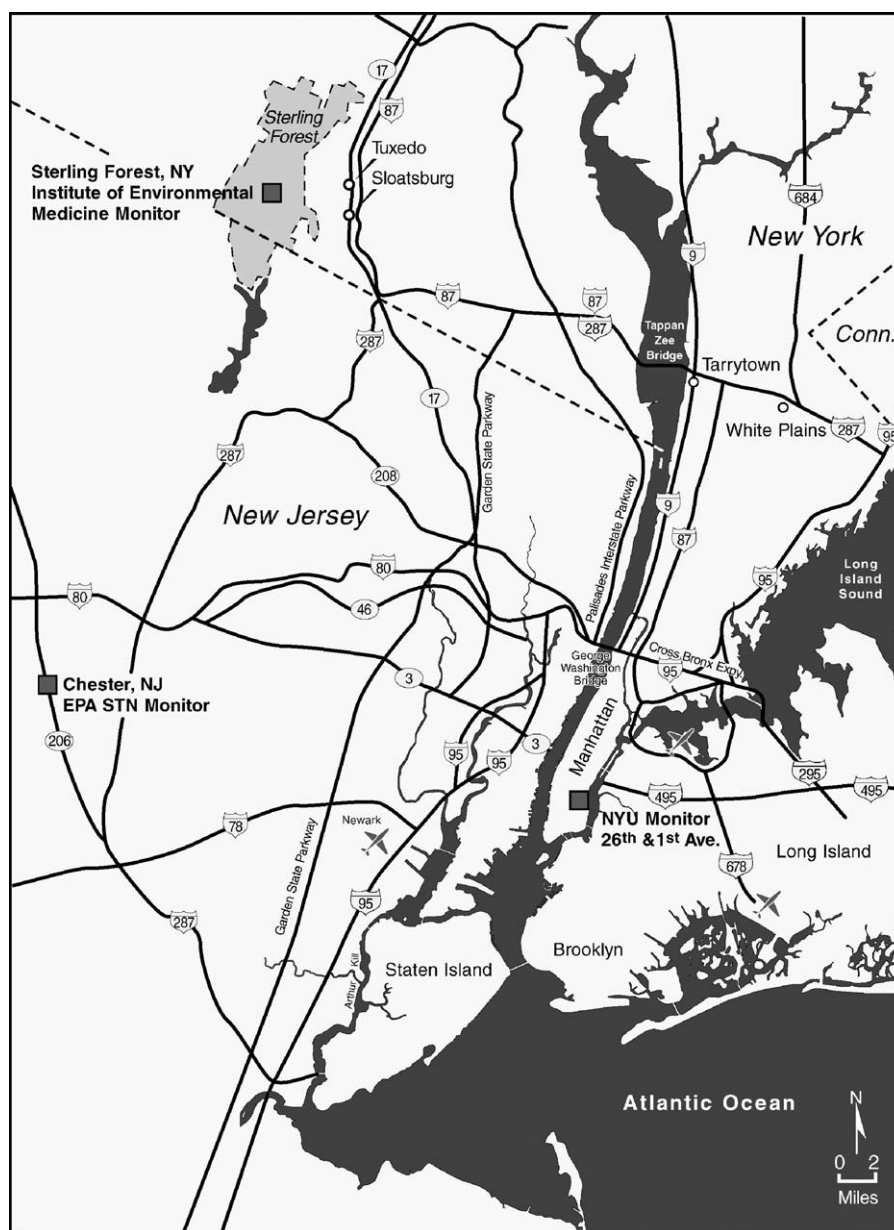


Fig. 1. Location of the NYU and EPA STN Monitors.

Table 1
Number of observations used in the comparison between two sites (n), and the correlation of $PM_{2.5}$, sulfur, OC and EC

	N	$PM_{2.5}$	n	S	OC	EC
SF vs. CH	88	0.75	50	0.95	0.69	0.33
HC vs. CH	87	0.80	52	0.96	0.74	0.52
SF vs. HC	312	0.82	312	0.92	0.70	0.49

SF = Sterling Forest (NY), NYC = New York City (NY) and CH = Chester (NJ).

combustion-related sources. Similar conclusions have been made by another study (Schwab et al., 2004). The slight EC correlation found among the sites are likely related to the three locations having similar local weather conditions from day to day. NYC, Sterling Forest and Chester are therefore impacted similarly by regional/transported aerosols (also see Fig. 2). The impact of transported sulfur is especially clear from the trajectory map of the five highest and five lowest sulfur days experienced in

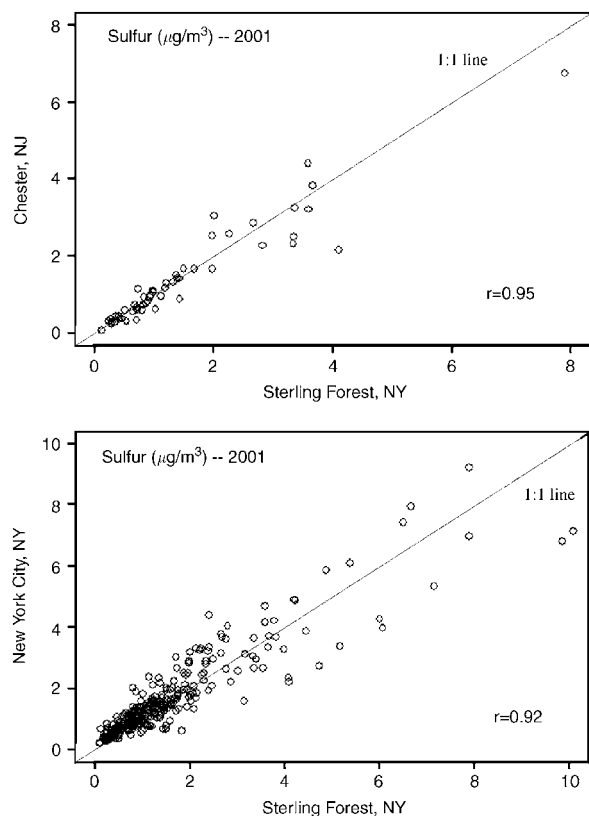


Fig. 2. Sulfur measurements collected at the two background sites, Sterling Forest, NY and Chester, NJ; and, sulfur measurements collected at Sterling Forest versus New York City, NY.

NYC (Fig. 3). These 72-h back trajectories were computed using NOAA's online (internet-based) HYSPLIT model (Draxler and Rolph, 2003). The 5 highest sulfur (concentrations greater than $5 \mu\text{g m}^{-3}$) days are associated with westerly winds, which is consistent with other studies of the impact of transported aerosols in the Northeast; while the 5 lowest sulfur (less than $0.5 \mu\text{g m}^{-3}$) days are associated with winds from the N–NW. On the other hand, local NYC sources do not appear to affect Sterling Forest, given that on less than 4% of the days in 2001 winds were found to be blowing from NYC and towards Sterling Forest, i.e. from the SE direction. These facts validate the use of Sterling Forest as a reference site for our work. A summary of the $\text{PM}_{2.5}$, elemental and carbon concentrations for Sterling Forest and NYC are provided in Table 2.

Our data show that Sterling Forest is also useful in identifying contributions from trans-continental and other (non-sulfate related) transported dust

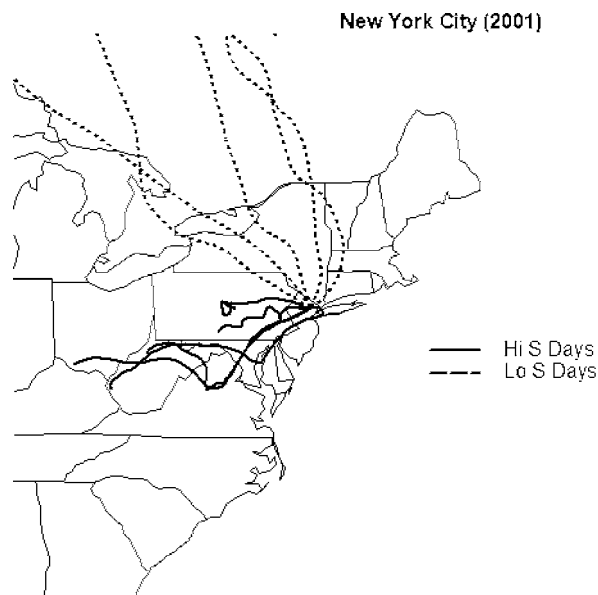


Fig. 3. 72h back-trajectories using NOAA's HYSPLIT model for the 5 highest and lowest sulfur days in NYC.

Table 2

Means and standard deviations of the $\text{PM}_{2.5}$ and elemental data for the 2 NYU sites (ng m^{-3})

	Sterling Forest (SF)		New York City (NYC)	
	Mean	Std. deviation	Mean	Std. Deviation
$\text{PM}_{2.5}$	11358	8301	17326	9642
Na	39	44	85	67
Mg	12	15	20	16
Al	42	57	44	50
Si	67	126	134	166
S	1512	1432	1583	1317
Cl	–2	11	37	273
K	36	27	50	49
Ca	20	19	60	36
Ti	4	18	4	4
V	3	3	10	6
Mn	1	1	7	11
Fe	39	40	194	131
Ni	4	14	24	14
Cu	1	2	6	13
Zn	9	7	44	97
Se	1	1	3	2
Br	3	9	8	37
Sr	1	1	3	3
Ba	0	3	9	5
Pb	2	5	9	36
OC	1700	856	3597	1260
EC	174	139	1226	580

episodes. Fig. 4 shows a plot of silica (Si) concentrations at the two monitoring sites, and the two clearly identifiable peaks at both sites at the

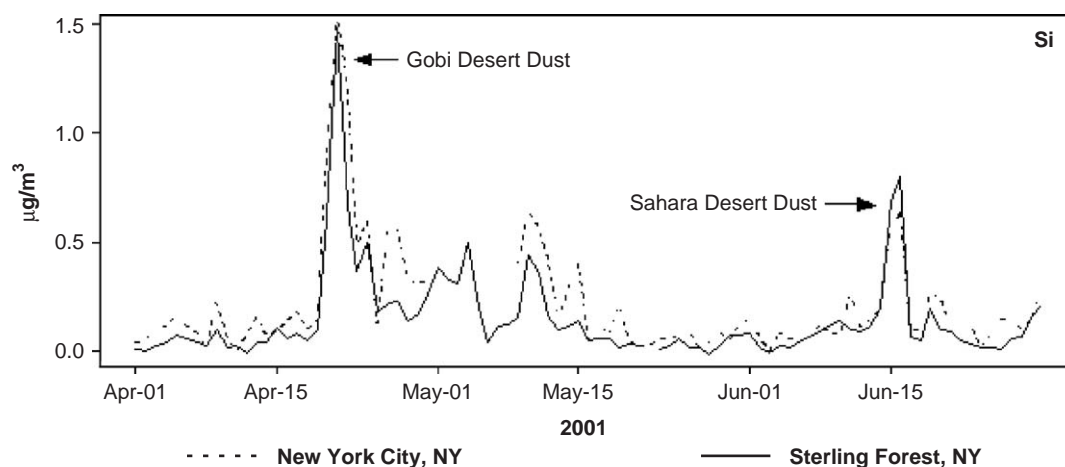


Fig. 4. Comparison of silica between the two NY sites during trans-continental desert dust episodes.

same time are clearly as a result of transported desert dust particles being carried across from the Gobi desert and the Sahara desert at those times. The unusual peak recorded at both our sites in April is the result of a well-documented aerosol dust plume originating in a Gobi Desert dust storm. This pollution was then transported across the Pacific Ocean to the West Coast of the US, and then on across the nation. This episode was well documented by the US EPA (US EPA, 2003). This trans-continental long-range transport of Gobi Desert dust is further confirmed by our data, showing major identical peaks at both our sites in April 2001 (Fig. 4).

In mid-June, a smaller regional dust peak is again observed at both our sites. Sequential satellite images taken at the period document a large plume originating in the Sahara Desert in Africa that migrated westward across the Atlantic Ocean in mid-June, 2001 (NASA, 2001). The episode recorded at our site on 15–16 June is due to an early precursor of the larger plume that mainly impacted the Caribbean and the Gulf of Mexico a few days later (19–21 June 2001). Backward wind-trajectories for NYC for 15–16 June confirm winds blowing from over the Atlantic Ocean to NYC, suggesting that the winds might have intercepted the earlier parts of this large desert dust plume. Examination of satellite photos also confirm an early pulse of aerosol into the mid-Atlantic at that time. STN data for NYC do not reveal this episode, as samples were not collected on a daily basis by NYC STN sites, and it so happened that no STN samples were scheduled for collection on these particular days

(15–16 June 2001). These two aerosol episodes clearly demonstrate that transported aerosols are similarly experienced at both the Sterling Forest and NYC sites under consideration in this research.

4. PMF source apportionments

Case 1: PMF analysis separating transported and local sulfur.

This source apportionment analysis of NYC data (that incorporates the Sterling Forest sulfur) identifies six sources of $PM_{2.5}$, as shown in Table 3. The two sulfur variables considered in the NYC source apportionment analysis now clearly defines sources as being either transported or local. Two of the sources, namely, “transported sulfates” and “transported desert dust” are from non-local sources. On the other hand, “residual oil burning”, “traffic”, “local dust” and “WTC” are clearly local NYC pollution sources.

Time-series plot of the mass contributions from each source are provided in Fig. 5. Total NYC $PM_{2.5}$ mass concentrations were regressed onto the six factors to translate factor scores into these mass contributions from each source:

$$NYC PM_{2.5} = \beta_0 + \beta_1 * G_{TRANS.} + \beta_2 * G_{TRAFFIC} \\ + \beta_3 * G_{RESIDUAL OIL} + \beta_4 * G_{SOIL} \\ + \beta_5 * G_{LOCAL DUST} + \beta_6 * G_{WTC},$$

where G are the factor scores of the component, and the β 's are the regression coefficients from the regression of mass onto the factor scores.

Table 3

Factor loadings (correlations) and the annual and seasonal mass contributions ($\mu\text{g m}^{-3}$) for 2001 (95% CI of contribution estimates) from the six sources identified for NYC using two Sulfur variables

	Transp. sulfates	Traffic	Residual oil	Transp. desert dust	“Local” dust	WTC
Na	0.18	−0.06	−0.07	0.37	0.41	0.18
Mg	0.13	0.09	−0.15	0.81	0.21	0.06
Al	0.11	0.12	−0.23	0.88	0.15	−0.18
Si	0.11	0.09	−0.12	0.88	0.19	0.29
Cl	−0.08	−0.08	0.07	−0.08	−0.04	0.91
K	0.15	0.08	−0.09	0.34	0.17	0.76
Ca	0.11	0.40	−0.14	0.78	0.35	0.09
Ti	0.13	0.11	−0.22	0.83	0.28	0.00
V	0.45	0.37	0.40	0.13	0.18	0.06
Mn	0.06	0.15	−0.16	0.02	0.99	0.05
Fe	0.12	0.36	−0.16	0.37	0.87	0.06
Ni	−0.09	0.01	0.90	−0.18	−0.13	−0.08
Cu	0.03	0.11	−0.04	−0.03	0.17	0.95
Zn	−0.02	0.08	0.03	−0.04	0.09	0.93
Se	0.58	0.40	0.09	−0.01	0.17	0.15
Br	−0.01	−0.08	−0.04	−0.03	−0.02	0.98
Sr	−0.02	−0.14	0.10	0.14	−0.08	0.72
Ba	0.23	0.43	−0.04	0.53	0.14	−0.02
Pb	−0.02	−0.03	0.01	−0.04	0.04	0.99
OC	0.65	0.82	−0.32	0.19	0.32	0.10
EC	0.39	0.80	0.20	0.10	0.17	0.10
S (TRANSP.)	0.92	0.44	−0.25	0.11	0.01	−0.02
S (LOCAL)	−0.20	0.12	0.09	−0.01	0.11	0.10
Eigen value	2.21	2.42	1.43	4.31	2.53	5.86
2001 annual contributions (UG/M3)	7.9 (7.4–8.4)	6.7 (5.9–7.5)	3.4 (3.0–3.9)	1.1 (0.8–1.4)	0.4 (0.1–0.6)	0.4 (0.4–0.5)
Winter (JAN–MAR ‘01)	6.4 (5.9–6.8)	4.8 (4.2–5.3)	7.7 (6.7–8.7)	0.5 (0.3–0.6)	0.2 (0.1–0.3)	0.3 (0.3–0.4)
Summer (JUN–AUG ‘01)	12.6 (11.8–13.4)	8.8 (7.8–9.8)	0.9 (0.8–1.1)	1.1 (0.8–1.4)	0.4 (0.1–0.6)	0.1 (0.1–0.2)

The “transported sulfates” source has high correlations with sulfur, as well as with OC and selenium, indicating that other secondary aerosols are included in this component, and that much of this pollution is derived from coal burning, a major source of selenium in the eastern US. This source has a strong seasonal pattern, with higher levels in the summertime. The “transported desert dust” component profile has high percentages of elements associated with the earth’s crust (e.g., Al and Si). This “source” has two very prominent peaks which as mentioned previously are attributed to dust transported as a result of desert storms in the Gobi and Sahara deserts. However, there appears to be small contributions year-round, which cannot be attributed to transport. Since elements associated with these storms are similar to those measured in NYC on a daily basis, both local and non-local contributions have aligned on the same “source”. However, since levels associated with the dust storms are much higher than usual (more than 5

times higher than normal: Gobi = $6.6 \mu\text{g m}^{-3}$, and Sahara = $5.1 \mu\text{g m}^{-3}$), this factor has been categorized as transported aerosols.

Among the “local” NYC sources, “traffic” and “residual oil” are found to be the significant contributors. However, in terms of mass contributions these levels are much lower than those associated with the “transported sulfates”. The “traffic” component is a mixture of elemental carbon and organics from vehicular exhaust; and, traces of re-suspended road dust including elements like Fe and Ca. As expected, this component has a strong day of week pattern with much lower concentrations found on the weekends (mean = $3.8 \mu\text{g m}^{-3}$) compared to weekdays (mean = $7.8 \mu\text{g m}^{-3}$), confirming its identification as traffic-related (as traffic in NYC is much heavier on weekdays). The lowest 5 percentile of traffic contributions ($< 0.9 \mu\text{g m}^{-3}$) coincided with Sundays mostly, and few with Saturdays and holidays (Christmas and 12/26). The “Residual Oil” component’s elemental profile has nickel and vanadium, two

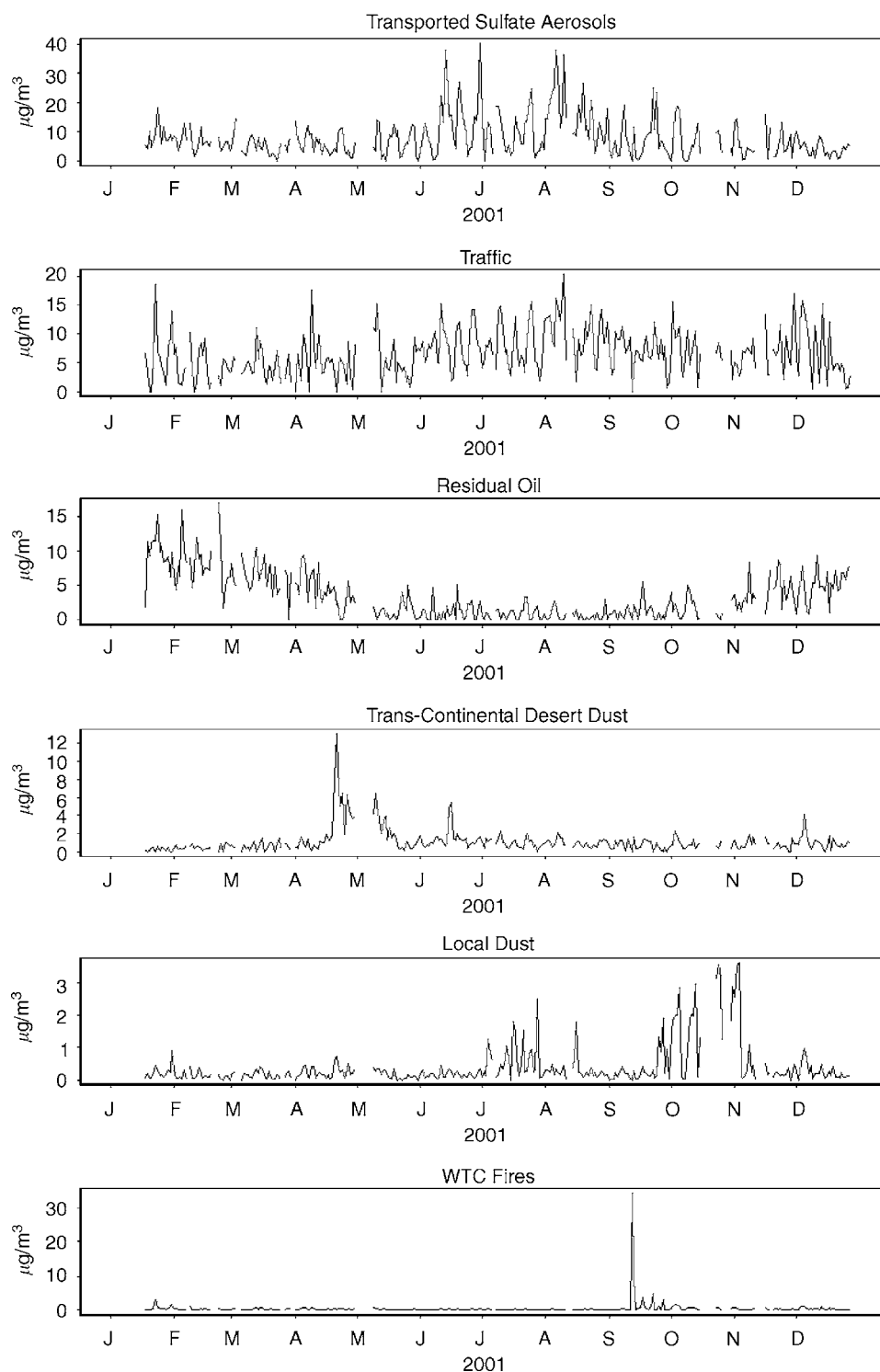


Fig. 5. Time-series plots of the PM_{2.5} contributions ($\mu\text{g}/\text{m}^3$) from the six source categories identified for NYC.

tracers commonly associated with this source. The time-series plot of this source also depicts a characteristic seasonal pattern, where colder months are

associated with higher contributions, as a result of greater demand for space heating during winter (winter mean = $7.7 \mu\text{g m}^{-3}$ vs. summer mean = $0.9 \mu\text{g m}^{-3}$).

As expected, this source is found to be highly correlated with EPA's SO₂ gaseous data ($r = 0.58$). The "WTC" plume is a result of the destruction and fires following the attack on the WTC towers and this plume laden with chlorine, zinc, lead, copper and potassium particles hit our Manhattan site on the evening of 12 September. The iron–manganese source most likely is a result of re-suspended dust that is local in origin, and is therefore labeled as "local dust". This source constitutes for less than half a $\mu\text{g m}^{-3}$ for 2001 and an increase in levels are found in the dryer months, from July to early August and again for a period between late September and mid-November. These elevated levels towards the latter part of the year might include the re-suspension of WTC-related dust during the Ground Zero cleanup in October–November 2001. Overall, the PM_{2.5} was fit well by the model (slope = 1.0, R -squared = 0.87).

In this analysis, the transported aerosols together constitute more than 40% of the total annual PM_{2.5} mass measured at NYC. Of this, the "transported sulfates" are the majority, and this classification is found to have the greatest mass impact of all the individual source categories, either local or non-local. There is a notable seasonal variation in this component, with higher levels being observed in the summer, most likely as a result of photochemistry. During the summer months, as much as 56% of the total PM_{2.5} can be attributed to transported sulfates. Wind trajectories on the highest days from this source, mostly indicate this source as being from the West and around the Ohio region where coal fired power plants are still being operated.

Case 2: Separate PMF analyses of transported and local NYC concentrations.

Table 4

Factor loadings (correlations) and the seasonal and annual 2001 source PM_{2.5} mass contributions ($\mu\text{g m}^{-3}$) from the two transported sources and the four local sources identified for NYC using Sterling Forest (SF) data and the differences in concentrations between SF and NYC elemental data

	Transported		Local			
	Transp. aerosols	Transp. desert dust	Residual oil	Traffic	"Local" dust	WTC
Na	0.06	0.43	−0.23	0.12	0.36	0.17
Mg	0.06	0.76	−0.19	0.34	0.17	0.05
Al	0.07	0.84	−0.17	0.31	0.12	−0.19
Si	0.07	0.86	−0.13	0.34	0.15	0.28
S	0.92	0.23	−0.06	0.43	0.02	0.01
Cl	−0.06	−0.05	−0.02	−0.06	−0.05	0.92
K	0.12	0.37	−0.10	0.19	0.16	0.75
Ca	0.11	0.68	−0.04	0.59	0.35	0.07
Ti	0.08	0.81	−0.20	0.29	0.25	−0.01
V	0.33	0.20	0.41	0.45	0.14	0.05
Mn	0.03	0.15	−0.15	0.01	0.97	0.02
Fe	0.08	0.44	−0.12	0.34	0.85	0.03
Co	−0.02	0.02	0.65	0.11	−0.11	0.03
Ni	−0.08	−0.15	0.90	0.02	−0.13	−0.07
Cu	0.03	0.02	−0.07	0.08	0.18	0.93
Zn	0.01	0.02	0.02	0.05	0.10	0.93
Se	0.48	0.10	0.20	0.37	0.14	0.13
Br	0.00	0.00	−0.07	−0.05	−0.02	0.98
Sr	−0.02	0.14	−0.06	0.06	−0.12	0.72
Ba	0.22	0.51	0.00	0.60	0.11	−0.02
Pb	−0.01	0.00	−0.03	−0.01	0.04	0.99
OC	0.65	0.29	−0.09	0.67	0.29	0.09
EC	0.39	0.18	0.33	0.78	0.13	0.09
Eigen value	1.88	4.20	1.80	2.90	2.31	5.79
2001 Annual contributions (UG/M3)	7.0 (6.5–7.5)	1.0 (0.6–1.5)	3.8 (3.1–4.5)	4.6 (3.7–5.4)	0.7 (0.4–1.0)	0.4 (0.3–0.5)
Winter (JAN–MAR '01)	5.0 (4.7–5.4)	0.6 (0.4–0.9)	6.1 (4.9–7.2)	3.8 (3.1–4.6)	0.2 (0.1–0.4)	0.3 (0.2–0.4)
Summer (JUN–AUG '01)	12.4 (11.6–13.3)	1.1 (0.7–1.6)	2.5 (2.0–2.9)	5.3 (4.3–6.3)	0.7 (0.4–0.9)	0.1 (0.1–0.2)

Table 5

Fraction of total PM_{2.5} mass and elemental/carbon concentrations as a result of transported aerosols in NYC

	I. PMF analysis incorporating 2 sulfur variables: S(SF) & S(NYC-SF)			II. Two separate PMF analyses of SF and “local” (NYC-SF) data sets		
	Annual (2001)	Winter (Jan–Mar ‘01)	Summer (Jun–Aug ‘01)	Annual (2001)	Winter (Jan–Mar ‘01)	Summer (Jun–Aug ‘01)
PM _{2.5}	0.44	0.33	0.56	0.45	0.34	0.61
S	0.80	0.79	0.85	0.90	0.86	0.94
OC	0.31	0.26	0.67	0.40	0.32	0.53
EC	0.12	0.08	0.15	0.05	0.04	0.09

In the second approach (case 2), two independent source apportionment analyses were conducted: one conducted on the Sterling Forest (SF) elemental concentrations, and a separate analysis of the NYC “local” elemental concentration variables, as derived from the differencing of NYC–SF elemental concentrations. The source apportionment analysis of Sterling Forest data identifies three components. Two out of the three are clearly identifiable transported sources; namely, “Transported Sulfates” and “Trans- Continental Dust”. The third component was harder to identify with this one year of data, and this component did not match with any of the components identified when PMF was applied to the raw NYC data, so this component was therefore left out of the subsequent NYC transported vs. local source apportionments. To the extent to which this unidentifiable Sterling Forest source component actually contributes to the NYC site (if it does), the reported analysis would understate the transport contribution to the NYC aerosol. Thus, our estimated transported contributions may be overly conservative (i.e. underestimating transport).

In the second source apportionment (for case 2), the calculated differences in elemental concentrations between the two sites were used to resolve four “local” sources: “residual oil”, “traffic”, “WTC” and “local dust” (see Table 4). For the purpose of this analysis, we included the two-clear-cut transported components from the Sterling Forest PMF analysis along with the four local components into the NYC source apportionment:

$$\text{NYC PM}_{2.5} = \beta_0 + \beta_1 * G_{\text{SULPHATES (T)}} + \beta_2 * G_{\text{SOIL (T)}} \\ + \beta_3 * G_{\text{TRAF (L)}} + \beta_4 * G_{\text{RESID OIL (L)}} \\ + \beta_5 * G_{\text{LOC DUST (L)}} + \beta_6 * G_{\text{WTC (L)}},$$

where G are the factor scores for transported (T) and local (L) components; and the β 's are the

regression coefficients from the regression of mass onto the factor scores

Overall, the PM_{2.5} was also fit well by this model (slope = 1.0, R -squared = 0.81).

Table 5 provides the estimates of the fraction of PM_{2.5}, sulfur, organic carbon and elemental carbon that is transported into New York City on an annual and seasonal basis, for each of the approaches applied in this study. The fractions are based on the attributed mass and elemental concentrations obtained from each of the analysis.

5. Discussion

The major role of aerosols transported into NYC indicated by our analysis highlights the significance of the upwind sources on the impact of the city's air quality. As mentioned previously, this transport phenomenon has been well documented by several past studies that identify transported aerosols as a major part of PM problem in the northeastern US. Some of these past studies have made estimations as to the contributions of transported aerosols in NYC through external site-to-site comparisons of individual elements that are assumed to serve as tracers of transported aerosols (e.g., S). For example, the New York Summer Aerosol Study, conducted almost 30 years ago, found that, on certain summer study days, most of the sulfate and a significant fraction of total suspended particulate matter (TSP) were due to long-range transport from sources in the mid-west by comparison with rural sites (Lippmann and Kneip, 1979). At that time, based on back-trajectory analysis and sulfate data for multiple sites, the study attributed an estimated 73% of measured NYC sulfate to transport.

A more recent study suggests that only one-half of the sulfate levels in Queens are attributable to transport (Dutkiewicz et al., 2003). This estimate

was based on using a single-site analysis using variations in sulfate as a function of the wind direction at this site to quantify local and transported sulfates. Sulfur levels for each of the wind direction sectors were computed, with the authors assuming that the wind sector with the lowest sulfur levels indicated the local contributions to the total sulfate. Based on this assumption, a constant “local” sulfur concentrations was determined, and this was then subtracted from the total sulfur levels from each of the other (higher S) wind quadrants to estimate the sulfate fraction from transported aerosols at that site. However, it is possible that even the lowest wind direction sulfur also included some transported sulfur. Thus, this assumption could have potentially resulted in an overestimation of local sulfur, and the subsequent underestimation of the transported fraction. Thus, it may be that studies that assume all sulfate is transported overestimate transport, while analyses that assume the lowest direction sulfate is all of local origins will underestimate transport.

Source apportionment studies are important for addressing these concerns by helping us to better understand all the different sources contributing to PM mass, thereby allowing an attribution of this transport source along with other local sources. Recently, two source apportionment studies were conducted using STN data collected at sites in NYC (Ito et al., 2004; Qin et al., 2006). In both studies, separate and independent source apportionments were conducted for each of the sites. Therefore, at each site, a set of sources were resolved, and most sources were found to be common to the multiple sites. Among the common sources was a “secondary sulfate” source that the studies suggest is due to the transport of aerosols. This is one way of assessing the impact of transported secondary sulfates. However, as the source apportionment has been conducted for each individual site, and there is no unique tracer for this transport “source”, so one cannot confidently assign all the mass from this factor to transport (as local impacts may also be incorporated). As a result, many past source apportionment studies, including the two other NYC studies, have resolved a “secondary sulfates” component, but such a definition is still ambiguous as to what part of it could be attributed to transport. In contrast, our approach is able to more definitively assign the sulfate source to transport as it uses sulfur measurements from a background site that like NYC experiences transported aerosols

similarly. Being able to less ambiguously assign PM mass to local vs. transported contributions is especially important for a thorough assessment of the air quality issues in NYC, which can thereby allow the implementation of more effective pollution controls and better aid the regulatory decision-making process.

In order to provide additional insight into our use of this differing approach, we have also conducted a “regular” PMF source apportionment analysis of just the NYC elemental dataset, without incorporating the SF data (results not presented here). This conventional approach actually provided results qualitatively and quantitatively similar to those found for Case 1 (i.e. resolving similar sources and comparable source contributions). These “conventional PMF” apportionment results were also similar to those reported in the work of Ito et al., (2004) and Qin et al. (2006) for other NYC sites. However, our Cases 1 and 2 analyses results, by first subtracting off the transported component, provided greater certainty that the sulfate component identified in our analyses of NYC data was, indeed, due to non-local aerosols transported into NYC.

Slight, but informative, differences in annual mass estimations are also found when comparing the respective apportionments from our Cases 1 and 2 approaches. Seasonal differences were more pronounced in the first approach, especially for traffic and residual oil. Subtracting the concentrations between both sites for ALL the elements might not be the more suitable option. Organic Carbon measurements at Sterling Forest might also consist of local production of OC by trees. Considering these factors, it appears that taking a difference in OC between the two sites (i.e., Case 2) might also have the side effect of underestimating local NYC OC levels. This, in turn, could cause some underestimation of local sources. Therefore, in the case of NYC, our first and simpler approach (Case 1: using the SF sulfur tracer of transported aerosol), may be the slightly preferable approach of the two we applied.

Two major local sources, traffic and residual oil, along with transported sulfates are found to also have significant impacts on NYC’s air quality on a regular basis. This is consistent with other recent NYC source apportionment studies (Ito et al., 2004; Qin et al., 2005). On 25% of the days in 2001, transported sulfates, traffic and residual oil combustion were greater than 10, 9 and $5\mu\text{g m}^{-3}$, respectively. Transported sulfates and traffic appear to

have higher contributions in summer than in winter, while residual oil is the largest contributor to $\text{PM}_{2.5}$ in the winter months. In comparison, local dust and transported desert dust only intermittently have discernable effects on the $\text{PM}_{2.5}$ concentration, with each of their annual contributions being less than $1 \mu\text{g m}^{-3}$. Emissions from ground zero were identified and found to last for a few weeks. However, the worst impact from this source was experienced on 12 September with 24-h average concentration from this single source being greater than $30 \mu\text{g m}^{-3}$ on that day.

Finally, the single greatest advantage of factor-analysis based receptor models (such as PMF) clearly illustrated in this work is the fact that, using such an approach, no quantitative knowledge of the types or elemental profiles of sources is required in order to identify even very unusual sources. Instead, using the ambient elemental concentration data supplied, the model seeks to independently explain most of the variability among the elements in terms of a smaller set of “sources”. The validity of this process is especially apparent in this analysis due to the fact that the PMF was able to identify and quantify two very clear and well documented aerosol pollution episodes as distinct $\text{PM}_{2.5}$ sources: the WTC fires’ plume impact in mid-Manhattan on 12 September 2001, and the Gobi Desert and Sahara Desert sandstorm inter-continental dust transport episodes in April and June 2001, respectively.

Overall, our results suggest that transported aerosols have a major impact on NYC’s air quality, despite improvements in regional air pollution since the imposition of the 1970 Clean Air Act. In fact, nearly half the total $\text{PM}_{2.5}$ reported in NYC is attributable to transport into the city on annual basis, and more than half (nearly two-thirds) of the $\text{PM}_{2.5}$ in the summertime. For sulfate, the transported percentage is much greater, reaching approximately 90% of all the sulfate impacting the site considered in downtown Manhattan. Therefore, roughly half of the city’s $\text{PM}_{2.5}$ problem is outside the control of NYC regulation, and in order for the city to be in compliance with current standards, the cleaning up of local sources alone may not be sufficient to meet ambient standards in all locations.

This is the first study to use the process of subtracting elemental mass from a reference site prior to its statistical source apportionment analysis as a means to more clearly separate local vs. non-local sources. Air pollution at urban sites is a complex mixture resulting from aerosols produced

locally, as well as those formed in the atmosphere and/or transported from great distances. This greatly complicates the discernment of sources, especially when much of the mass is transported regionally. Having additional independent data from a nearby reference site (unaffected by local sources) as indicator of transported pollution is helpful in more clearly identifying local sources and, in turn, in distinguishing the local vs. transported source components in this complex mixture.

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